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Pesticides as a Waste Problem with Examples from Norway

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1. Introduction

Pesticides can produce waste problems when disposing off the surplus from usage, containers and other residues, but also during the production and distribution. Obsolete pesticides such as the chlorinated γ-HCH (lindane) and DDT have created huge problems also because the use of toxic raw materials, such as chlorophenols and dioxins, and the production of hazardous and useless by-products form the production, such as e.g. α- and β- HCH that constitutes about 85% of the production output. The cleanup of production sites and dumps has created major problems - the work is almost completed in Western Europe - but many locations remain polluted, especially in Central- and Eastern Europe, and possibly also in other sites around the globe. Sites polluted with obsolete pesticides are mainly threatening rural areas. According to the World Health Organization (WHO) half of the deaths due to cancer in 2050 will be caused by polluted food, soil, water and environment, mainly from pesticides. The economical burden will also be heavy. The scale of the problem with obsolete pesticides is estimated to more than 250 000 tons of pesticides in Easter Europe and Central Asia (Vijgen & Egenhofer, 2009).

Landfilling is the most common waste treatment worldwide. In The European Union (EU) several waste directives have improved the quality of the waste going to landfills, both containing hazardous waste and ordinary household or municipal solid waste (MSW), but as we show here there is a potential for pollution from pesticides both from old and new landfills. As outlined in the Waste Framework Directive from the EU pesticides are one of the key parameters that needs to be monitored. Based on the results from Norwegian investigations on pesticides from greenhouse production an increased focus is put on this problem in Europe.

Large quantities of numerous chemicals have the potential to pollute the waste that is landfilled. Annually close to 500 kg is landfilled for each person in the developed world, producing on the average more than 30 tons of waste over a lifetime. Before a more managed waste disposal started in the nineties many landfills were established and scattered almost everywhere, with the risk of polluting the environment and threatening human health. Landfills create polluted wastewater, or leachate, ca. 50-200 mm annually in dry areas, 400-800 mm on the average, and >1000 mm under wet conditions. The content of organic matter in the waste is often the most important factor influencing the quality of the leachate, in addition to the local geology, hydrology and climate. Landfills usually create large emissions of organic matter and nitrogen, see Figure 1, measured as chemical oxygen demand (COD), biochemical oxygen demand (BOD) and ammonia-nitrogen (NH_4-N). Nitrogen is usually the parameter with the longest half-life. Also the leachate contains suspended solids including organic and inorganic colloids, estimated to more than 12 kg/day and 2.5 kg/day, respectively, from an average landfill. These particles also have a strong potential to erode and transport pesticides from the waste, especially the hydrophobic ones with low water solubility and often high toxicity. The COD and BOD contain mainly organic acids in leachate from fresh waste. Later the BOD is reduced, and the COD will consist mainly of "hard" COD due to non-degradable humic substances.

Pesticides are labelled as hazardous and are generally not allowed to be landfilled. Pesticides that are resisting anaerobic degradation and have a high water solubility and a low acid partition coefficient, $pK_a<7$, are expected to be more readily leached from the waste. Herbicides generally have a high solubility, in the order of several hundreds or thousands of mg/l, fungicides have intermediate solubility, up to a few mg/l, while insecticides are usually non-soluble in water, with water solubility less than 10 μg/l. The solubility is, however, depending on factors such as the content of dissolved organic carbon, e.g. for DDT (Haarstad & Fresvig, 2000). Landfill leachate has high concentrations of suspended and soluble organic matter and thus has a potential of transporting relatively large amounts of hydrophobic compounds. Leachate is generally anaerobic and methanogenic with a high pH, or acidic and low in pH if the waste is fresh, or a combination of these. The ecotoxicity of the pesticides are usually inversely related to their solubility, but the solubility is often much greater than the ecotoxicological limit value (the predicted no effect concentration or PNEC).

The production and use of pesticides clearly are able to produce waste problems that can give environmental and health problems. It is necessary to impose strict regulations both on the production, sale, use and waste handling of these compounds, all based on intensive monitoring.

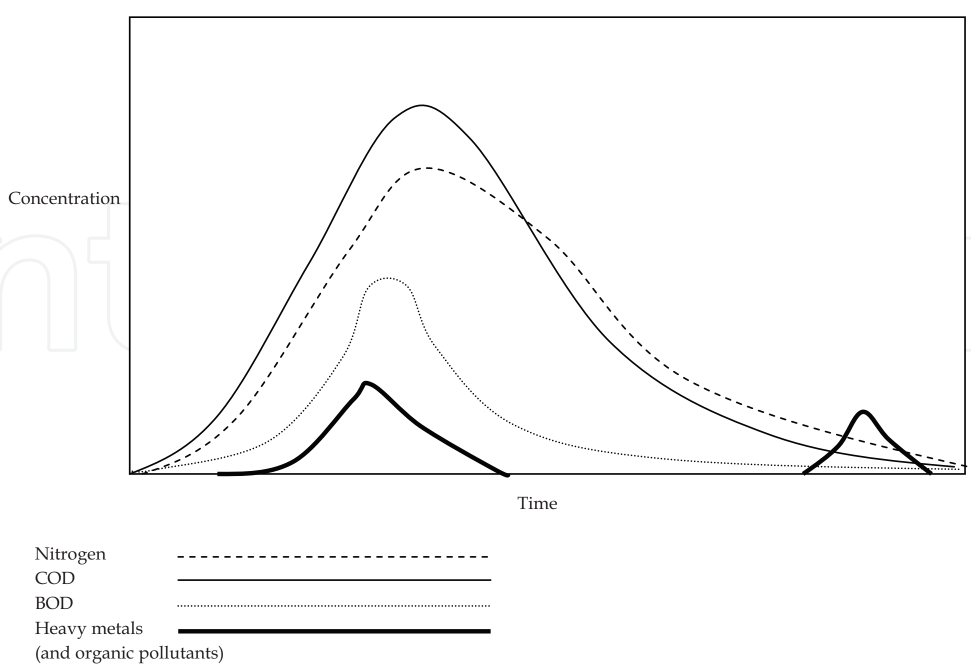

Fig. 1. Trends in concentrations (mg/l or μg/l) in leachate emissions over a lifetime of a landfill.

2. Pesticides in ordinary landfills

There are limited reports on the concentrations of pesticides in MSW leachate. A review listed 24 compounds, including 3 metabolites, found in concentrations ranging from 0.025 to 26 μg/l (chlorprofam) - most of the findings were however from one sampling (Kjeldsen et al., 2008). Another study found phenoxy acids in concentrations up to 65 μg/l in leachate-contaminated ground water (Tuxen et al., 2003). A report showed the occurrence of mecoprop and isoproturon in UK leachates, of which the former was readily removed by aerobic treatment (Robinson & Barr, 1999). In a study pesticides were analyzed in 28 grab samples of untreated and 12 samples of treated leachate from eight municipal solid waste (MSW) landfills, over a period of 10 years, with the purpose to screen for compounds and to evaluate the removal in different treatment systems (Haarstad & Mæhlum, 2008). Also 1 grab sample of leachate sediments is included. The most frequently detected group of compounds are the phenoxy acids, and they also occur in highest concentrations, up to 230 μg/l for mecoprop, in this study. Also three fungicides and one insecticide were detected, but in much lower concentrations. All samples exceeded the maximum limit value (MLV) for the sum concentration of pesticides in drinking water (0.5 μg/l), and six compounds exceeded the PNEC. Reverse osmosis showed good removal of phenoxy acids, while sequential batch reactor aerobic treatment, as well as aerated lagoons in combination with wetlands, groundwater infiltration and reactor treatment showed slightly lower removal.

Table 1 shows that most pesticides are detected in leachate treated in wetlands, mostly with pre-areation (Haarstad & Mæhlum, 2008), where the concentrations can reach 50 ppb for the most water soluble compounds. A total of 13 compounds were detected.

	Reverse osmosis	Aeration/wetlands	SBR
Pesticides in treated leachate	phenoxy acids	phenoxy acids	phenoxy acids
		chlorfenvinphos	
		isoptroutron	
		azoxystrobin	
		clopyralid	
		mecoprop	
Concentrations (μg/l)	0.01-0.08	0.16-50	0.03-1.1

Table 1. Pesticides detected in MSW leachate (from Haarstad & Mæhlum, 2008).

3. Pesticides in green waste from agriculture

Pesticides in agricultural waste products are also a problem. A landfill consisting mainly of organic waste from a tree nursery and containing an estimated 900 kg of DDT has been monitored since 1994. Downstream ground water was sampled from four wells. More than 10 years of monitoring of two of the wells is presented in Table 2, in addition to sampling of the waste. A total of seven pesticides were detected in the ground water (Haarstad, 2008). In addition to DDT, there were two other insecticides and four fungicides occurring in the ground water downstream of the landfill. The maximum concentration of pesticides was 3.76 µg/L of which 2.70 µg/L was permethrin in October 2000. The maximum concentration of DDT in the ground water was 0.52 µg/L, indicating that the leachate DDT concentrations probably exceed the water solubility of the compound. The investigation shows that the practice of establishing local landfills of waste from nurseries is environmentally unsafe. In 2002, the landfill was covered with clayey soil and vegetated. It seems that this has stabilized the pesticide concentrations in the ground water and removed the occurrence of extreme values. However, the vegetated soil layer has not been able to prevent the leakage of pesticides to the ground water, still present 5 years later.

	Well P2								Well P3					
Sampling date	Σ*	D	F	P	L	PE	I	T	Σ	D	L	P	PE	F
04.11.1994									0.12	0.12				
10.04.1995									0.15	0.15				
16.04.1996									0.07	0.07				
27.05.1998	0.32		0.07	0.25			0		0.3	0.22	0.08			
01.12.1998	0.28		0.15				0.13		0.89	0.47	0.33			0.09
11.08.1999									0.34	0	0.34			
10.12.1999	0.14			0.14					0.07	0.07				
16.10.2000	0.77		0.16			0.61			3.76	0.52	0.54		2.7	
26.11.2001	0.25	0.1	0.06	0.06	0.03				0.63	0.14	0.47			0.02
26.05.2003	0.14	0.03	0.11						0.76	0.11	0.65			
03.10.2003									0.94	0.08	0.86			
05.11.2004	0.11		0.02	0.09					0.27	0.04	0.18	0.04		0.01
26.06.2007	0.29							0.29	0.16	0.02	0.12	0.02		

From Haarstad 2008. Blank=below the limit of detection. Σ =sum pesticides. D=sum DDT, F=fenpropimorph, P=propiconazole, L=lindane, PE=permethrin, I=iprodione, T=trifloxystrobin

Table 2. Pesticides from green waste (µg/l)

4. Pesticides in products, waste and wastewater from greenhouse production

In a study of a greenhouse waste tip a total of 8 pesticides were detected with concentrations from 10 – 170 µg/kg. In the runoff receiving leachate from the tip concentrations of pesticides varied between 0.03 – 1. 2 µg/l.

In a study of runoff from greenhouses water samples downstream 9 large facilities focusing on flower production were analyzed for 56 pesticides. The samples included concentrated runoff from the production of tomatoes and cucumbers. Pesticides were detected at all locations and in 90% of the samples, with a total of 18 compounds, of which were 9 fungicides, 5 herbicides and 4 insecticides in concentrations exceeding the PNEC value. Detected fungicides were cyprodinil, propikonazol, iprodion, azoksystrobin, prokloraz and vinklozolin. Insecticides with detections above the PNEC were pirimikarb, diazinon and klorfenvinfos. The fungicides pyrimetanil, iprodion and imazalil had concentrations >1µg/l in the concentrated runoff from vegetable greenhouses. The findings where the concentrations were above the PNEC values came from two locations with the production of flowers, in addition to water from the floor of one of the greenhouses. Generally the most likely pesticides to leak from greenhouse productions are pyrimethanil, cyprodinil, propiconazole, iprodione, azoxystrobin, imazil, prochloraz and pirimicarb, based on the facts that they are common in use in such productions, results from other monitoring programs, timing of their detections downstream greenhouses and a correlation between findings and high levels of nutrients in the samples.

Repeated sampling of imported and domestic flowers, pot plants and pot soil shows the presence of occasional high levels of residual pesticides. In the pot plants 22 compounds were detected with concentrations varying from 0.02 to 8.8 mg/kg, while in pot soil 7 compounds were detected with concentrations from 0.05 to 2.2 mg/kg. Most detection was made in flowers, where 31 compounds were detected, with concentrations from 0.01 to 5.3 mg/kg. For pot plants 11 fungicides and 11 insecticides were detected, for pot soil 4 insecticides and 3 fungicides, and for flowers 14 insecticides and 18 fungicides.

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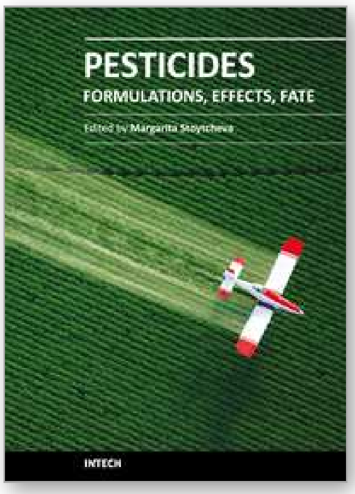

Pesticides - Formulations, Effects, Fate
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This book provides an overview on a large variety of pesticide-related topics, organized in three sections. The first part is dedicated to the "safer" pesticides derived from natural materials, the design and the optimization of pesticides formulations, and the techniques for pesticides application. The second part is intended to demonstrate the agricultural products, environmental and biota pesticides contamination and the impacts of the pesticides presence on the ecosystems. The third part presents current investigations of the naturally occurring pesticides degradation phenomena, the environmental effects of the break down products, and different approaches to pesticides residues treatment. Written by leading experts in their respective areas, the book is highly recommended to the professionals, interested in pesticides issues.

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